

# Microscopic theory of spin-triplet $f$ -wave pairing in quasi-1D organic superconductors

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We present a microscopic theory of fluctuation-mediated pairing mechanism in organic superconductors (TMTSF)<sub>2</sub>X, where the experimentally observed coexistence of  $2k_F$  charge fluctuation and  $2k_F$  spin fluctuation is naturally taken into account. We have studied, within the random phase approximation, the extended Hubbard model at quarter-filling on a quasi-one-dimensional lattice, where we consider the off-site repulsive interaction up to third next nearest neighbors along with the on-site repulsion. The results show that spin-triplet  $f$ -wave-like pairing can be realized in this system, dominating over singlet  $d$ -wave-like pairing, if  $2k_F$  spin and  $2k_F$  charge fluctuations coexist.

It has been a long standing issue to clarify the superconducting state of quasi-one-dimensional (q1D) organic superconductors (TMTSF)<sub>2</sub>X ( $X = \text{PF}_6, \text{ClO}_4$ , etc.), so called the Bechgaard salts.<sup>1,2</sup> One may expect unconventional superconducting states due to the quasi-one-dimensional nature of these materials as well as the electron correlation effects generally seen in organic materials. In fact, there have been experiments suggesting the existence of nodes<sup>3,4,5</sup>, while recent experiments showing a large  $H_{c2}$ <sup>6,7</sup> and an unchanged Knight shift across  $T_c$ <sup>3,8</sup> supports a realization of spin-triplet pairing. The pairing symmetry of (TMTSF)<sub>2</sub>X has thus become quite a hot issue very recently.

Theoretically, triplet  $p$ -wave pairing state in which the nodes of the pair potential do not intersect the Fermi surface has been proposed from the early days<sup>9,10,11</sup>. However, the occurrence of triplet pairing in (TMTSF)<sub>2</sub>X is curious from a microscopic point of view since superconductivity lies right next to a  $2k_F$  spin density wave (SDW) in the pressure-temperature phase diagram<sup>12</sup>. Naively, SDW spin fluctuations should favor spin-singlet  $d$ -wave-like pairing as suggested by several other authors<sup>13,14,15</sup>. However, one should note that the insulating phase is not pure SDW at least for some anions, namely,  $2k_F$  charge density wave (CDW) actually coexists with  $2k_F$  SDW.<sup>16,17</sup> In fact, one of the present authors has phenomenologically proposed<sup>20</sup> that triplet  $f$ -wave-like (see Fig.1(c) for a typical pair potential) pairing may dominate over  $p$ -wave pairing and become competitive against  $d$ -wave-like pairing (see Fig.1(b)) due to a combination of quasi-1D (disconnected) Fermi surface and the coexistence of  $2k_F$  SDW and  $2k_F$  CDW fluctuations. A similar scenario has been proposed by Fuseya *et al.*<sup>21</sup> Concerning the  $f$ -wave vs.  $d$ -wave competition, it has also been proposed that magneto tunneling spectroscopy<sup>22</sup> via Andreev resonant states<sup>23</sup> is a promising method to detect the  $f$ -wave pairing.

However, there has been no *microscopic* theory for  $f$ -wave pairing in (TMTSF)<sub>2</sub>X starting from a Hamiltonian that assumes only purely electronic repulsive interactions.<sup>24</sup> To resolve this issue, here we study an extended Hubbard model at quarter-filling on a quasi-one-dimensional lattice within the random phase approximation (RPA). We consider off-site repulsions up to third

nearest neighbors along with the on-site repulsion in order to naturally take into account the coexistence of  $2k_F$  charge and  $2k_F$  spin fluctuations. The merit of adopting RPA<sup>25</sup> is that we can easily take into account the off-site repulsion as compared to fluctuation exchange (FLEX) approximation, where it is by no means easy to take into account distant interactions<sup>26,27</sup>.

The model Hamiltonian is given as

$$H = - \sum_{\langle \mathbf{i}, \mathbf{j} \rangle, \sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_{|\mathbf{i}-\mathbf{j}|=m_l} V_{i,j} n_i n_j,$$

where  $c_{i\sigma}^\dagger$  creates a hole (note that (TMTSF)<sub>2</sub>X is actually a  $3/4$  filling system in the electron picture) with spin  $\sigma = \uparrow, \downarrow$  at site  $\mathbf{i} = (i_a, i_b)$ . Here,  $\langle \mathbf{i}, \mathbf{j} \rangle$  stands for the summation over nearest neighbor pairs of sites. As for the hopping parameters, we take  $t_{ij} = t_a$  for nearest neighbor in the (most conductive)  $a$ -direction, and  $t_{ij} = t_b$  for nearest neighbor in the  $b$ -direction. We choose  $t_b = 0.2t_a$  to take into account the quasi-one-dimensionality.  $t_a$  is taken as the unit of energy throughout the study.  $U$  and  $V_{ij}$  are the on-site and the off-site repulsive interactions, respectively. We take distant off-site repulsions because it has been shown previously that nearest neighbor and second nearest neighbor off-site repulsion is necessary to have coexistence of  $2k_F$  spin and  $2k_F$  charge density waves.<sup>18,19</sup> Here we take off-site repulsions up to third nearest neighbors, namely,  $V_{i,j} = V_0, V_1$  and  $V_2$  with  $m = |\mathbf{i} - \mathbf{j}| = 1, 2$  and  $3$ , respectively. The effect of the third nearest neighbor repulsion,  $V_2$ , will be discussed at the end of the paper. The effective pairing interactions for the singlet and triplet channels due to spin and charge fluctuations are given as

$$V^s(\mathbf{q}, \omega_l) = U + V(\mathbf{q}) + \frac{3}{2}U^2\chi_s(\mathbf{q}, \omega_l) - \frac{1}{2}(U + 2V(\mathbf{q}))^2\chi_c(\mathbf{q}, \omega_l) \quad (1)$$

$$V^t(\mathbf{q}, \omega_l) = V(\mathbf{q}) - \frac{1}{2}U^2\chi_s(\mathbf{q}, \omega_l) - \frac{1}{2}(U + 2V(\mathbf{q}))^2\chi_c(\mathbf{q}, \omega_l) \quad (2)$$

within RPA, where

$$V(\mathbf{q}) = 2V_0 \cos q_x + 2V_1 \cos(2q_x) + 2V_2 \cos(3q_x) \quad (3)$$

and  $\omega_l$  is the Matsubara frequency. Here,  $\chi_s$  and  $\chi_c$  are the spin and charge susceptibilities, respectively, which are given as

$$\chi_s(\mathbf{q}, \omega_l) = \frac{\chi_0(\mathbf{q}, \omega_l)}{1 - U\chi_0(\mathbf{q}, \omega_l)} \quad (4)$$

$$\chi_c(\mathbf{q}, \omega_l) = \frac{\chi_0(\mathbf{q}, \omega_l)}{1 + (U + 2V(\mathbf{q}))\chi_0(\mathbf{q}, \omega_l)}. \quad (5)$$

Here  $\chi_0$  is the bare susceptibility given by

$$\chi_0(\mathbf{q}, \omega_l) = \frac{1}{N} \sum_{\mathbf{p}} \frac{f(\epsilon_{\mathbf{p}+\mathbf{q}}) - f(\epsilon_{\mathbf{p}})}{\omega_l - (\epsilon_{\mathbf{p}+\mathbf{q}} - \epsilon_{\mathbf{p}})}$$

with  $\epsilon_{\mathbf{k}} = -2t_a \cos k_a - 2t_b \cos k_b - \mu$  and  $f(\epsilon_{\mathbf{p}}) = 1/(\exp(\epsilon_{\mathbf{p}}/T) + 1)$ .  $\chi_0$  peaks at the nesting vector  $\mathbf{Q}$  ( $= (\pi/2, \pi)$  here) of the Fermi surface. The terms proportional to  $\chi_s$  and  $\chi_c$  in eqs. (1) and (2) represent effective pairing interactions due to the spin and charge fluctuations, respectively. The chemical potential  $\mu$  is determined so that the band is quarter-filled, which is  $\mu = -1.38$  here. We take  $U = 1.6$  throughout the study, which is large enough to have strong  $2k_F$  spin fluctuations (large  $\chi_s(\mathbf{Q}) = \chi_s(\mathbf{Q}, 0)$ ) but not so large as to drive SDW instability at high temperatures. The off-site interactions are chosen so that  $2k_F$  charge fluctuations are induced as discussed later. In the actual numerical calculation, we take  $N = 400 \times 40$   $k$ -point meshes except for low temperatures, where we take  $N = 800 \times 80$  meshes.

To obtain the onset of the superconducting state, we solve the gap equation within the weak-coupling theory,

$$\lambda \Delta(\mathbf{k}) = - \sum_{\mathbf{k}'} V^{s,t}(\mathbf{k} - \mathbf{k}', 0) \frac{\tanh(\beta \epsilon_{\mathbf{k}'}/2)}{2\epsilon_{\mathbf{k}'}} \Delta(\mathbf{k}'). \quad (6)$$

The transition temperature  $T_C$  is determined by the condition,  $\lambda = 1$ . In the weak coupling theory,  $\omega$  dependence of the pair potential  $\Delta(\mathbf{k})$  is neglected. Although this approximation is quantitatively insufficient, it is expected to be valid for studying the pairing symmetry of  $\Delta(\mathbf{k})$  mediated by both spin and charge fluctuations. In the following calculations, we study triplet and singlet cases with  $\Delta(\mathbf{k}) = -\Delta(-\mathbf{k})$  and  $\Delta(\mathbf{k}) = \Delta(-\mathbf{k})$ , respectively. We define  $\phi_s(\mathbf{k}) = \Delta(\mathbf{k})/\Delta_M$  and  $\phi_t(\mathbf{k}) = \Delta(\mathbf{k})/\Delta_M$  for singlet and triplet pairing, respectively, where  $\Delta_M$  is the maximum value of the pair potential.

From eqs.(1)-(5), it can be seen that when  $U \sim -(U + 2V(\mathbf{Q}))$  is satisfied,  $|V^s(\mathbf{Q})| \sim |V^t(\mathbf{Q})|$  holds, apart from the first order terms, which are negligible in the limit of strong spin and/or charge fluctuations (but turn out to be important in the actual cases considered later). This,

along with the disconnectivity of the Fermi surface (note that the number of nodes intersecting the Fermi surface is the same between  $f$ - and  $d$ -waves), is expected to make spin triplet  $f$ -wave pairing competitive against singlet  $d$ -wave pairing.<sup>20</sup>

We now move on to the calculation results. First, we focus on the case where spin fluctuation is dominant, *e.g.*,  $V_0 = V_1 = V_2 = 0$ . As shown in Fig. 1, the magnitude of  $\lambda$  for the singlet case is much larger than that for the triplet case. The resulting singlet pair potential  $\phi_s(\mathbf{k})$  changes sign as  $+-+-$  along the Fermi surface (see Fig. 1(b)). We call this  $d$ -wave pairing, where  $\phi_s(\mathbf{k})$  is roughly proportional to  $\cos(2k_x)$ . On the other hand, the triplet pair potential  $\phi_t(\mathbf{k})$  changes sign as  $+-+-$  along the Fermi surface (see Fig. 1(c)). We call this  $f$ -wave, where  $\phi_t(\mathbf{k})$  is roughly proportional to  $\sin 4k_x$ . The results here are expected from the previous FLEX study.<sup>20</sup>

Let us now move on to the cases where we turn on the off-site repulsions. In order to have the coexistence of  $2k_F$  spin and  $2k_F$  charge fluctuations as experimentally observed, namely, to have  $\chi_s(\mathbf{Q}) \sim -\chi_c(\mathbf{Q}) = -\chi_c(\mathbf{Q}, 0)$ ,  $U \sim -(U + 2V(\mathbf{Q}))$  has to be satisfied as mentioned earlier. To accomplish this,  $V_1$  has to be close to  $U/2$ , as can be seen from eq.(3). Namely, since the  $x$  component of  $\mathbf{Q}$  is  $Q_x = \pi/2$ , the  $V_1$  term in eq.(3) is dominant for  $q \simeq \mathbf{Q}$ , making  $U \sim -(U + 2V(\mathbf{Q}))$  if  $V_1 \simeq U/2$ . Thus we first choose  $V_1 = U/2 = 0.8$ . As for the other off-site repulsions, we first choose  $V_0 = 1.2$  and  $V_2 = 0.5$  as a typical value. As shown in Fig. 2(a) and 2(b),  $\chi_s$  and  $\chi_c$  peak around  $(k_a, k_b) = \pm(\pi/2, \pi), \pm(\pi/2, -\pi)$  and the maximum values are both about 9.1, so that we indeed have the situation where  $2k_F$  spin and  $2k_F$  charge fluctuations coexist. As shown in Fig. 2(c), the magnitude of  $\lambda$  for triplet pairing is now much larger than that for singlet pairing. The corresponding singlet pair potential  $\phi_s(\mathbf{k})$  has the  $d$ -wave form as shown in Fig.2(d), while the triplet one  $\phi_t(\mathbf{k})$  has the  $f$ -wave form as shown in Fig.2(e). Note that the result of  $\lambda_{\text{triplet}} \gg \lambda_{\text{singlet}}$  is rather unexpected from the previous phenomenological argument<sup>20</sup> because when  $\chi_c(\mathbf{Q}) = \chi_s(\mathbf{Q})$ ,  $f$ -wave is only *degenerate* with  $d$ -wave in the previous theory. The origin of this discrepancy is the first order terms in eqs.(1) and (2), which are neglected in the phenomenological theory. Thus, we have obtained a remarkable result here, namely,  $f$ -wave can *completely* dominate over  $d$ -wave when  $2k_F$  spin and charge fluctuations coexist.

In order to further look into this point, we next reduce  $V_1$  from 0.8, thereby suppressing the charge fluctuation. The maximum value of  $\chi_c$  (not shown) is 5.2 and 3.2 for  $V_1 = 0.78$  and  $V_1 = 0.75$ , respectively. Although the maximum value of  $\chi_c$  is smaller than that of  $\chi_s$  ( $= 9.1$ ) in these cases,  $\lambda$  for the triplet case is still larger than ( $V_1 = 0.78$ ) or competitive against ( $V_1 = 0.75$ ) that for the singlet case as seen in Fig.3. This means that  $f$ -wave pairing has a chance to be realized *even if spin fluctuation dominates, as far as  $2k_F$  charge fluctuation exist to some extent*.<sup>20</sup>

Finally, in order to look into the effect of the third

nearest neighbor interaction  $V_2$ , we set  $V_2 = 0$  leaving the other parameters the same as in Fig.2.

As seen in Fig.4(a), the peak of  $\chi_c$  is slightly shifted towards  $4k_F$  ( $q_x = \pi$ ) when  $V_2 = 0$ , compared to the case of  $V_2 = 0.5t$  shown in Fig.2(a). ( $\chi_s$  is the same as Fig.2(a) since  $V_2$  does not affect  $\chi_s$  within the present formalism.) In this case, the singlet-triplet competition becomes much more subtle as seen in Fig.4(b). The reason for this can be found in Fig.4(c), namely,  $\phi_s(\mathbf{k})$  in this case has the same sign on most portion of the Fermi sur-

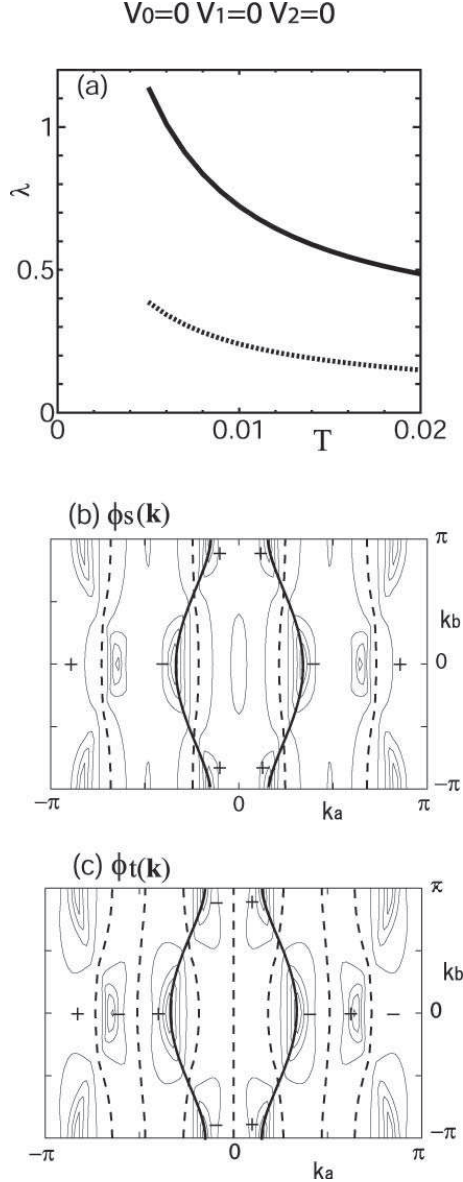


FIG. 1: Calculation results for  $U = 1.6$ ,  $V_0 = V_1 = V_2 = 0$ . (a) Temperature dependence of  $\lambda$  for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (b)  $\phi_s(\mathbf{k})$  and (c)  $\phi_t(\mathbf{k})$  at  $T = 0.01$ . In (b), and (c), the solid lines represent the Fermi surface and the dotted lines denote the nodal lines of the pair potentials.

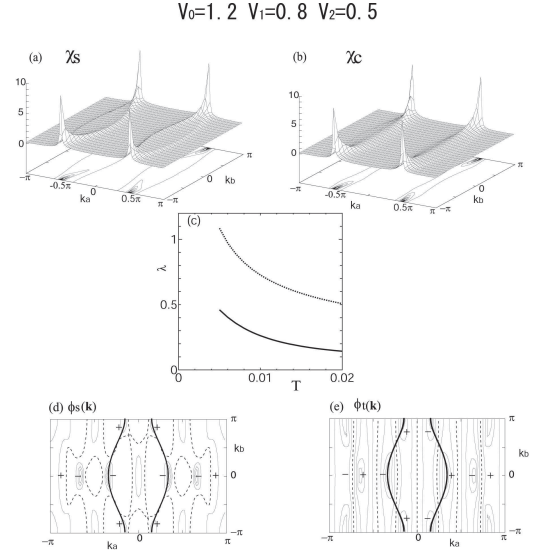


FIG. 2: Calculation results for  $U = 1.6$ ,  $V_0 = 1.2$ ,  $V_1 = 0.8$ ,  $V_2 = 0.5$ : (a)  $\chi_s(\mathbf{k})$  at  $T = 0.01$ . (b)  $\chi_c(\mathbf{k})$  at  $T = 0.01$ . (c) Temperature dependence of  $\lambda$  for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (d)  $\phi_s(\mathbf{k})$  and (e)  $\phi_t(\mathbf{k})$  at  $T = 0.01$ . In (d), and (e), the solid lines represent the Fermi surface and the dotted lines denote the nodes of the pair potentials.

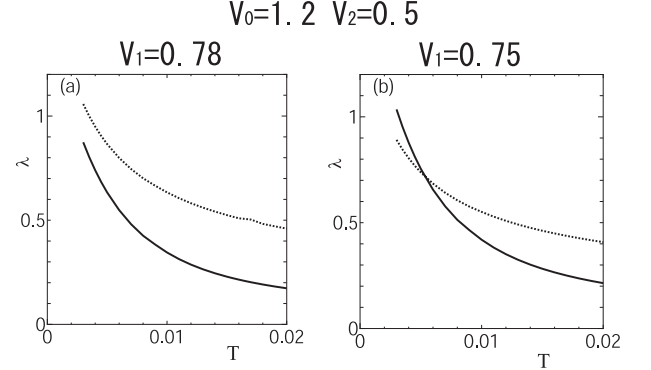


FIG. 3: Calculation results for  $U = 1.6$ ,  $V_0 = 1.2$ , and  $V_2 = 0.5$ . (a) Temperature dependence of  $\lambda$  for singlet (solid line) and triplet (dotted line) pairings for  $V_1 = 0.78$ . (b) Temperature dependence of  $\lambda$  for singlet (solid line) and triplet (dotted line) pairings for  $V_1 = 0.75$ .

face. In other words, it is more like *s*-wave than *d*-wave. This ‘*s*-wave’ pairing is induced by charge fluctuation, which does not totally cancel out with spin fluctuation in eq.(1) because the wave vector at which  $\chi_c$  peaks deviates from that for  $\chi_s$ . Since the ‘*s*-wave’ pair potential has the same sign on most portion of the Fermi surface, almost all the pair scattering processes on the Fermi surface, mediated by the *attractive* interaction (note the minus sign in eq.(1)) due to charge fluctuation, have positive contribution to superconductivity, making singlet pairing much more enhanced compared to the case with nonzero  $V_2$ .

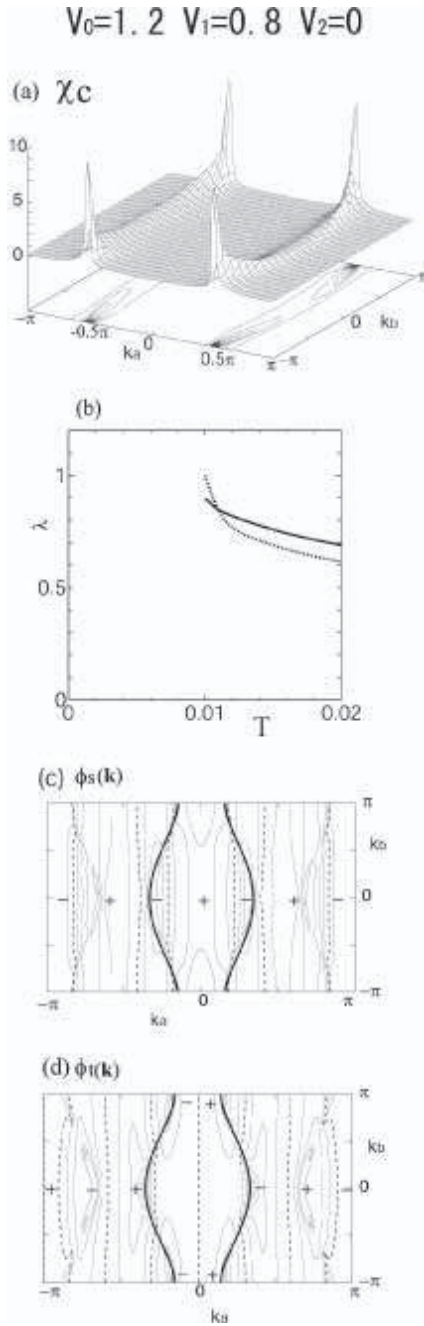


FIG. 4: Calculation results for  $U = 1.6$ ,  $V_0 = 1.2$ ,  $V_1 = 0.8$ ,  $V_2 = 0$ : (a)  $\chi_c(\mathbf{k})$  at  $T = 0.01$ . (b) Temperature dependence of  $\lambda$  for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (c)  $\phi_s(\mathbf{k})$  and (d)  $\phi_t(\mathbf{k})$  at  $T = 0.01$ . In (c) and (d), the solid lines represent the Fermi surface and the dotted lines denote the nodes of the pair potentials.

Conversely, the present results show that  $V_2$  has the effect of stabilizing  $2k_F$  charge fluctuation, which has a tendency to shift towards  $4k_F$  fluctuation when only  $V_0$  and  $V_1$  are present, and this effect in turn suppresses singlet pairing because in that case a strong cancellation occurs between the third and the fourth terms in eq.(1). Since the screening effect is known to be weak in quasi-one-dimensional systems, it is likely that such a distant off-site repulsion is present in the actual  $(\text{TMTSF})_2\text{X}$ .

To summarize, we have presented a microscopic theory of pairing mechanism in organic superconductors  $(\text{TMTSF})_2\text{X}$ , where we have taken into account the coexistence of the  $2k_F$  charge fluctuation and  $2k_F$  spin fluctuation by considering off-site repulsions up to third nearest neighbors. We have shown that the  $f$ -wave triplet pairing symmetry can be realized in this system when  $2k_F$  charge density fluctuation and  $2k_F$  spin density fluctuation coexists. Surprisingly, the condition for realizing  $f$ -wave pairing is much more eased compared to that in the previous phenomenological theory<sup>20</sup>.

In the present paper, we have neglected the realistic shape of the Fermi surface observed in the actual  $(\text{TMTSF})_2\text{X}$ <sup>28</sup>. Although the influence of this effect on the  $f$ -wave pairing is expected to be small because the  $x$ -component of the nesting vector is close to  $\pi/2$  in any case, a detailed analysis remains as a future study. Experimentally, it would be interesting to verify the realization of  $f$ -wave superconducting state by some phase sensitive probes.<sup>23,29</sup>

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